

Phase-reversed structures in superlattice of nonlinear materials

D. A. Antonosyan^{1,2,*} and G. Yu. Kryuchkian^{1,2,†}

¹*Yerevan State University, Alex Manoogian 1, 0025, Yerevan, Armenia*

²*Institute for Physical Researches, National Academy of Sciences,
Ashtarak-2, 0203, Ashtarak, Armenia*

We present detailed description of so-called phase-reversed structures that are characterized by two grating wave vectors allowing simultaneously phase-match two parametric three-wave processes. The novelty is that the structure is realized as a definite assembly of nonlinear segments leading to detailed description of cascaded three-photon processes with the parameters of realistic structured nonlinear materials of finite length. We apply these results for analysis of the quasi-phase-matching in production of both photon triplet and four-photon states in cascaded down-conversion. The received results are matched with the experimental data.

Keywords: Multi-photon processes, quasi-phase-matching, phase-reversed superlattice, down-conversion.

I. INTRODUCTION

Cascading processes in optical materials that involve several different second-order nonlinear interactions provide an efficient way to lower the critical power of optical parametric devices. The concept of multistep cascading brings new ideas into this field, leading to the possibility of an enhanced nonlinearity-induced phase shift, the simultaneous generation of higher-order harmonics, multi-color parametric interaction, difference-frequency generation, (see, [1] and references therein). Multistep parametric interactions and multistep cascading are characterized by at least two different nearly phase-matching or quasi-phase-matching (QPM) parameters. QPM is an important technique in nonlinear optics and is widespread used for various applications [1, 2]. The width of the phase-matching curve depends on the type of the crystal, its length, and the phase-matching method. QPM not only makes efficient frequency conversion possible, but also enables diverse applications such as beam and pulse shaping, multi-harmonic generation, high harmonic generation and all-optical processing. Recently, it has been demonstrated that the quantum-computational gates, multimode quantum interference of photons and preparation of multi-photon entangled states also can be realized

in QPM structures [3–6].

One of the non-uniform QPM structures is so-called phase-reversed structures [7], which has been studied both theoretically and experimentally and have been employed for the wavelength conversion, and for the cascaded single-crystal third harmonic generation process [8, 9]. Several proposals for increasing the phase-matching region have also been suggested, including the use of the phase-reversed QPM structures. These structures are based on the use of a phase-reversed sequence of many equivalent uniform short QPM domains, particularly, periodically poled domains, in which neighboring nonlinear segments have the opposite signs of the second-order susceptibility. The phase-reversed structures arranged in such a way that at the place of the joint two ends of the domains segments have the same sign of the $\chi^{(2)}$ coefficient (see Fig. 1). On the whole, this structure is characterized by two grating wave vectors allowing phase-match two processes. In the standard approach, the one-dimensional $\chi^{(2)}(z)$ function leading to the phase-reversed QPM structure can be described by the infinite Fourier expansion on two periods. Such phenomenological approach qualitatively explains the simultaneous QPM of two parametric nonlinear processes, however, has not included the parameters of realistic structured nonlinear materials. These dates often should be taken into to consideration if we are interest in more detailed description of cascaded parametric processes.

In this paper, we present more detailed description of phase-reversed structures in another way as the definite assembly of nonlinear segments. Then, we arrange this structure for two cascaded parametric devices proposed for generation of three-photon and four-photon states. In this way, we consider cascaded collinear parametric processes in one dimensional media. The paper is arranged as follows. In Sec. II we consider an effective nonlinear coefficient $G(\Delta k)$ of three-wave interaction in one-dimensional phase-reversed media. This quantity is defined as the Fourier transform of the second order susceptibility $\chi^{(2)}(z)$ in term of wave vectors mismatch Δk of three-wave interaction caused by dispersion in an optical material. In Sec.III we briefly consider application of

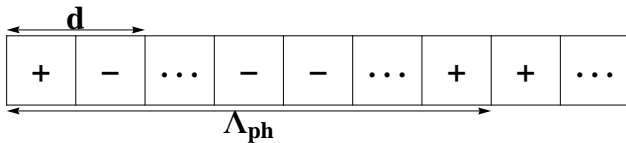


FIG. 1: Scheme of periodically poled nonlinear crystal (PPNC) which involves nonlinear domains of length l with $\chi_+ > 0$ and $\chi_- < 0$ with phase-reversal grating (with period Λ_{ph}) upon a uniform QPM grating (with period d).

*antonosyand@ysu.am

†kryuchkian@ysu.am

these results for obtaining QPM in generation of multi-photon states.

II. PHASE-REVERSED STRUCTURES OF FINITE LENGTH

We start with consideration of effective coupling constant $G(\Delta k)$ that usually describe three-wave interaction in second-order nonlinear media with the susceptibility $\chi^{(2)}(z)$ in the following form

$$G(\Delta k) = \int_0^L \chi^{(2)}(z) e^{i\Delta k(z)} dz, \quad (1)$$

where Δk is the phase-matching parameter and L is a length of medium. In the phenomenological approach the $\chi^{(2)}(z)$ function leading to the phase-reversed QPM structure can be described by the following Fourier expansion

$$\chi^{(2)}(z) = \chi_0 \sum_{n=-\infty}^{\infty} g_n e^{iG_n z} \sum_{m=-\infty}^{\infty} g_m e^{iF_m z}, \quad (2)$$

that is the product of two periodic functions. Here: $g_0 = 0$, $g_{n \neq 0} = 2/\pi n$, $g_{m \neq 0} = 2/\pi m$ and $G_n = \frac{2\pi}{d}n$, $F_m = \frac{2\pi}{\Lambda_{ph}}m$. In this case the effective interaction constant reads as

$$G(\Delta k) = \chi_0 L \sum_{n=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} g_n g_m e^{-i\frac{L}{2}(\Delta k - G_n - F_m)} \text{sinc}\left(\frac{L}{2}(\Delta k - G_n - F_m)\right). \quad (3)$$

Considering the phase-reversed QPM structure as the definite assembly of nonlinear segments we turn to the general formula obtained for the effective coupling constant in multilayered media [10]

$$G(\Delta k) = \sum_m l_m \chi_m e^{-i(\varphi_m + \frac{\Delta k_m l_m}{2})} \text{sinc}\left(\frac{\Delta k_m l_m}{2}\right),$$

$$\varphi_m = \sum_n^{m-1} l_n \Delta k_n, \quad \varphi_1 = 0, \quad (4)$$

where $\Delta k_n = k_0^n - k_1^n - k_2^n$ is the phase-matching function of three photon process in the each n -th domain,

$k_j^n(z, \omega) = \frac{\omega_j}{c} n_n(z, \omega_j)$, $n_n(\omega)$ is the refractive index of the medium at the given frequency.

In the result, the effective nonlinear coefficient $G(\Delta k)$ for the scheme depicted in the Fig.1 can be calculated in the following form

$$G(\Delta k) = L \chi_0 e^{-i\alpha(\Delta k)} Y_{M,N}(\Delta k). \quad (5)$$

We assume that phase-matching parameters are the same for all layers $\Delta k_n \equiv \Delta k$, as well as $\chi_+ = |\chi_-| \equiv \chi_0$.

We introduce the QPM function $Y_{M,N}(\Delta k)$ which is the product of the standard phase-matching spectral function for a single segment, i.e. $\text{sinc}(\frac{l\Delta k}{2})$, and the assemble function

$$Y_{M,N}(\Delta k) = \text{sinc}\left(\frac{l\Delta k}{2}\right) \frac{1}{MN} \frac{\sin\left[\frac{Nl}{2}(\Delta k - G)\right]}{\sin\left[\frac{l}{2}(\Delta k - G)\right]} \frac{\sin\left[\frac{MNI}{2}(\Delta k - F)\right]}{\sin\left[\frac{NI}{2}(\Delta k - F)\right]}. \quad (6)$$

Here: N -is the number of domains in each block, M -is the number of blocks, $\alpha(\Delta k) = \frac{Nl}{2}[(\Delta k - G) + (M - 1)(\Delta k - F)]$, l -is the length of each domain, $G = 2\pi/d$, $F = 2\pi/\Lambda_{ph}$, $d = 2l$, $\Lambda_{ph} = 2Nl$, $L = MNl$. It is easy to

check that the function $Y_{M,N}(\Delta k)$ has an alternative representation, as the product of expansions on the number of domains and blocks

$$Y_{M,N}(\Delta k) = \text{sinc}\left(\frac{l\Delta k}{2}\right) \frac{1}{MN} e^{i\varphi} \sum_{n=1}^N e^{-il(n\Delta k - G_n)} \sum_{m=1}^M e^{-iNI(m\Delta k - F_m)}, \quad (7)$$

where $\varphi = \frac{l}{2}(\Delta k - G)$. As we see, the above expressions (5), (7) derived on the base of the method of superlattices elaborated on the multilayered media differs from the formula (3), which is obtained in the standard, phenomenological approach. In contrast to the expression (3), the results (6),(7) depend on the number of the domains and blocks and also describe the case of small numbers of the segments. It is remarkable, that in this case the QPM function displays specific interference effects. Nevertheless, as shows our analysis some consequences of both (3) and (5) results, for $N \gg 1$, $M \gg 1$, coincide qualitatively.

Note, that single-phase-matched process of three-photon interaction in multilayered media is also described by an assembly function. For PPLN structure (see, one of the block in Fig.1 containing N layers) the phase-matching function is calculated as

$$Y_N(\Delta k) = \text{sinc}\left(\frac{l\Delta k}{2}\right) \frac{1}{N} \frac{\sin\left[\frac{Nl}{2}(\Delta k - G)\right]}{\sin\left[\frac{l}{2}(\Delta k - G)\right]}, \quad (8)$$

where Δk is the wave vectors mismatch of three-wave interaction. This function exhibits a structure of single narrow peaks as discussed in [11].

The obtained QPM function $Y_{M,N}(\Delta k)$ allows simultaneously phase-match two parametric processes which is achieved by using two grating vectors. In order to illustrate the possibility of the double phase-matching in such superlattice structure we present a detailed analysis of the function $Y_{M,N}(x)$ in dependence on the parameter $x = \frac{l\Delta k}{2}$ in the graphical form (see, Figs. 2, 3). The assembly function exhibits the structure of peaks that are separated by π . Unlike to the assembly function for single-phase-matched process, $Y_N(\Delta k)$ function has more complex structure, particularly, each group of the extremes consists of thin structure of twin-narrow peaks, separated by oscillating low maxima. The $\text{sinc}\left(\frac{l\Delta k}{2}\right)$ multiplier causes decreasing the amplitude of the consecutive peaks. The behavior of $Y_{M,N}(x)$ function is different for even and odd values of M . As depicted in Fig. 2 for even values of M the twin peaks have the opposite signs, while for odd values of M the both narrow peaks have the same sign (see Fig. 3). The existence of twin peaks and oscillation between them are the main peculiarities of this structure. These twin narrow peaks can be chosen to phase-match two parametric processes involved into the multistep-cascading.

Here we represent $Y_{M,N}(\Delta k)$ function for the small number of domains to exhibit direct correlation between number of domains and the width of phase-matching function (see, Fig. 4). It is easy to notice that with the decrease of number of domains and blocks the narrow peaks in the phase-matching function becomes wider, as well as the number of oscillation between twin peaks decreases. Besides this, the positions of the maxima of the phase-matching function becomes slightly shifted, although the periodicity of the $Y_{M,N}(x)$ function is remained the same.

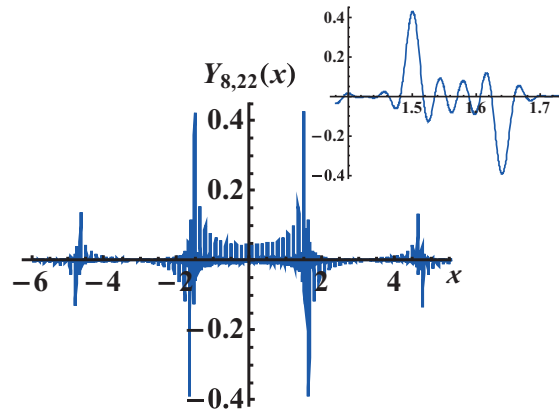


FIG. 2: Plot of the function $Y_{M,N}(x)$ for even numbers of both domains and blocks; $N = 22$ and $M = 8$. The inset shows the twin narrow peaks of opposite amplitudes.

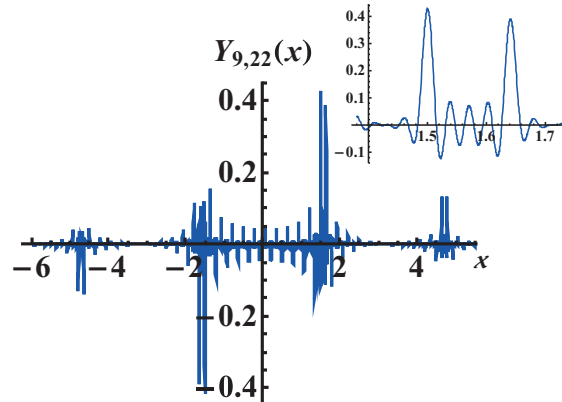


FIG. 3: Plot of the function $Y_{M,N}(x)$ for even numbers of $N = 22$ and odd numbers $M = 9$. The inset shows the twin narrow peaks of the same amplitudes.

III. APPLICATIONS: QPM FOR PRODUCTION OF MULIPHOTON STATES

In this section, we apply the above results analyzing the QPM in generation of both photon triplet and four-photon states in cascaded down-conversion. Recently, production of joint multiphoton states, particularly, photon triplet have attracted a great interest in probing the foundations of quantum theory and for various applications in quantum information technologies (see, [12–15] and the references therein). Depending on the configuration of the experiment and the designs of the crystal, the multiphoton states can be entangled in any number of variables: time, frequency, direction of propagation, and polarization. Studies carried out with multiphoton state beams range from the examination of quantum paradoxes, to applications in optical measurements, spectroscopy, imaging, and quantum information (see, [13, 16] and references there). In this way, the direct generation of photon triplets using cascaded photon-pairs has been

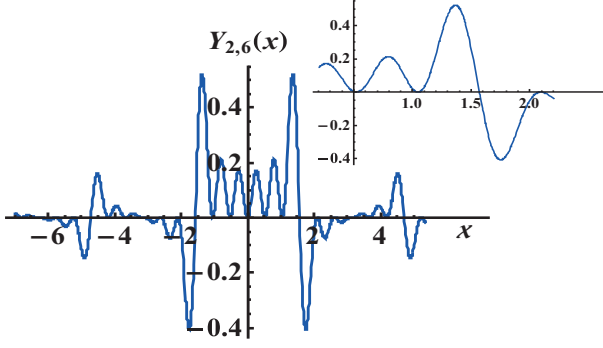


FIG. 4: Plot of the function $Y_{M,N}(x)$ for the small numbers of domains and blocks, namely $N = 6$ and $M = 2$. The insert shows the broadening of the peaks.

demonstrated in periodically poled lithium niobate crystals [14]. Most recently, joint quantum states of three-photons with arbitrary spectral characteristics have been studied on the base of optical superlattices [15]. In continuous of this paper, we analyze the cascaded generation of photon triplets in phase-reversed superlattice of $\chi^{(2)}$ materials.

We consider collinear, one-dimensional configurations focusing on consideration of cascaded parametric down-conversion (PDC), which generates three-photon states in the presence of an optical cavity. In the scheme considered the pump field at the frequency ω_0 converts to the subharmonics at the central frequencies $\omega_1 = \frac{\omega_0}{3}$ and $\omega_2 = \frac{2\omega_0}{3}$ throughout two cascaded processes: $\omega_0 \rightarrow \omega_1 + \omega_2$ and $\omega_2 \rightarrow \omega_1 + \omega_1$. Below we consider the amplitude of three-photon generation through the cascaded processes assuming the frequencies ω_1 and ω_2 as variable quantities distributed around the central values $\frac{\omega_0}{3}$ and $\frac{2\omega_0}{3}$, respectively. These processes are characterized by the wave-vector mismatches: $\Delta k_1 = k(\omega_0) - k(\omega_1) - k(\omega_2)$ and $\Delta k_2 = k(\omega_2) - k(\omega'_1) - k(\omega''_1)$. The model Hamiltonian for the system, in the rotating-wave approximation, is given by

$$H_{int} = i\hbar (E_0 e^{-i\omega_0 t} a_0^+ - E_0^* e^{i\omega_0 t} a_0) + i\hbar \zeta (a_0 a^+ b^+ - a_0^+ ab) + i\hbar \xi (a^{+2} b - a^2 b^+). \quad (9)$$

Here, ζ and ξ are the nonlinear coupling constants that are the Fourier transformations of the second-order nonlinear susceptibility $\chi^{(2)}(z)$ (see, Eq.(1)). The cavity modes are described by discrete creation and annihilation operators a^+, b^+ and a, b as E_0 describes the amplitude of the driving fields. For the case of the phase-reversed superlattice structure the constants of the processes $\omega_0 \rightarrow \omega_1 + \omega_2$ and $\omega_2 \rightarrow \omega_1 + \omega_1$ are calculated as

$$\begin{aligned} \zeta &\sim L e^{-i\alpha(\Delta k_1)} Y_{M,N}(x_1), \\ \xi &\sim L e^{-i\alpha(\Delta k_2)} Y_{M,N}(x_2), \end{aligned} \quad (10)$$

where $x_1 = l\Delta k_1/2$, $x_2 = l\Delta k_2/2$. In general, the amplitude of three-photons at the frequencies $\omega_1, \omega'_1, \omega''_1$ generated in cascaded down-conversion has been calculated in [15]. This quantity is proportional to the factor $E_0 \zeta \xi$. For the case of phase-reverse structure we can calculate three-photon amplitude in the following form

$$\Phi(\omega_1, \omega'_1, \omega''_1) = f(\omega_1, \omega_2) h(\Delta k_1, \Delta k_2), \quad (11)$$

where

$$h(\Delta k_1, \Delta k_2) = (MN)^2 Y_{M,N}(k_1) Y_{M,N}(k_2) \quad (12)$$

is the two-dimension assemble function that describes joint phase-matching, while the function $f(\omega_1, \omega_2) = Cl^2 \chi_0 E_0(\omega_0) e^{-i\alpha(\Delta k_1)} e^{-i\alpha(\Delta k_2)}$, depends on the parameters of three-wave interaction, the phase multipliers $\alpha(\Delta k_i) = \frac{Nl}{2}[(\Delta k_i - G) + (M-1)(\Delta k_i - F)]$ in the formula (5) and C is the normalization factor of three-photon amplitude. Three-photon down-conversion is controlled by energy conservation between the pump and daughter photons $\omega_0 = \omega_1 + \omega'_1 + \omega''_1$.

Analyzing the graphics of $Y_{M,N}(x)$ (see Fig. 3) in application to cascaded scheme of down-conversion we could arrange two twin peaks of the phase-matching function with the wave-vector mismatches of cascading processes. We demonstrate it using the experimental dates for the cascaded down-conversion [17]. For the typical value of pump wave length $\lambda_L = 0.53 \mu\text{m}$ and LiTaO_3 crystal mismatch parameters from experimental results are $\Delta k_1 = 0.32 \mu\text{m}^{-1}$ and $\Delta k_2 = 0.87 \mu\text{m}^{-1}$. We check that the simultaneous phase-matching of two processes is really realized on the formula (6) for the parameters: $l = 10.25 \mu\text{m}$, $N = 22$, $M = 9$. In this case, the Δk_1 match with the second peak of the first twin-peaks on the positive branch of the axes and the Δk_2 matches with the first peak of the second twin-peaks, which are situated next to the first twin-peaks and separated from them by the period. We got perfect correspondence of theoretical and experimental results.

The dependence of the spectral function $h(x_1, x_2)$ on the parameters x_1 and x_2 is shown in Figs. 5,6. As we see, the joint spectral function displays the various maxima in x_1, x_2 place that indicate the range of the results of the effective three-photon splitting and perfectly confirm the results exhibited in Fig. 2. Thus, the above results show that the efficient generation of three-photon subharmonic mode is possible in this scheme. The parametric gain of the three-photon down-conversion in the spectral range of the maxima of the function $h(x_1, x_2)$ is the result of the most simultaneous phase-matching of two cascading second-order processes.

In the end of the section we shortly discuss QPM for generation of four-photon states in cascaded parametric oscillator. This device consists of three intra-cavity modes. The pump mode driven by an external coherent driving field at the frequency ω_0 is converted into the pair of modes ω_2 , where $\omega_0 = \omega_2 + \omega_2 =$

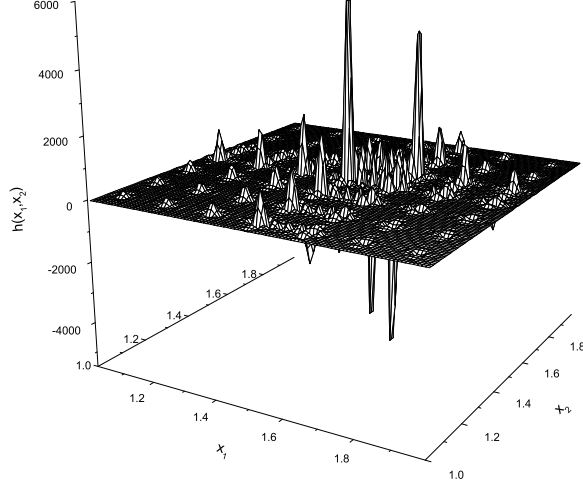


FIG. 5: The detailed plot of the function $h(x_1, x_2)$ in the range of double phase-matching peaks: $1 < x_1 < 2$ and $1 < x_2 < 2$, and the parameters $N = 22$, $M = 8$. There are four peaks: two positive, which correspond to $x_1 = 1.5$, $x_2 = 1.5$, $x_1 = 1.64$, $x_2 = 1.64$, and two negative at $x_1 = 1.5$, $x_2 = 1.64$ and $x_1 = 1.64$, $x_2 = 1.5$.

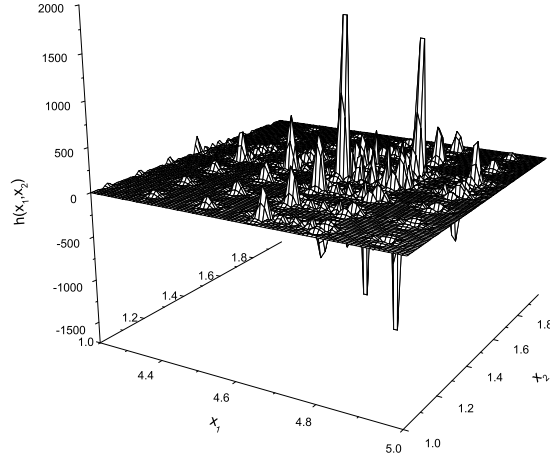


FIG. 6: Plot of the function $h(x_1, x_2)$ in the ranges $4.2 < x_1 < 5$ and $1 < x_2 < 2$, and the parameters $N = 22$, $M = 9$. The heights of the peaks are three times lower than for the peaks displayed on Fig. 5 in an agreement with the results presented on Fig. 2.

$\omega_0/2 + \omega_0/2$. Then, each of the mode ϖ_2 is transformed into second pair of modes ϖ_1 , where $\varpi_2 = \varpi_1 + \varpi_1 = \varpi_0/4 + \varpi_0/4$. These processes are characterized by the wave-vector mismatches: $\Delta k_1 = k(\varpi_0) - 2k(\varpi_2)$ and $\Delta k_2 = k(\varpi_2) - 2k(\varpi_1)$, respectively. To realize QPM, we use the typical value of four-photon down-conversion pump wave length $\lambda_p = 0,39\mu m$. In LiTaO_3 phase-

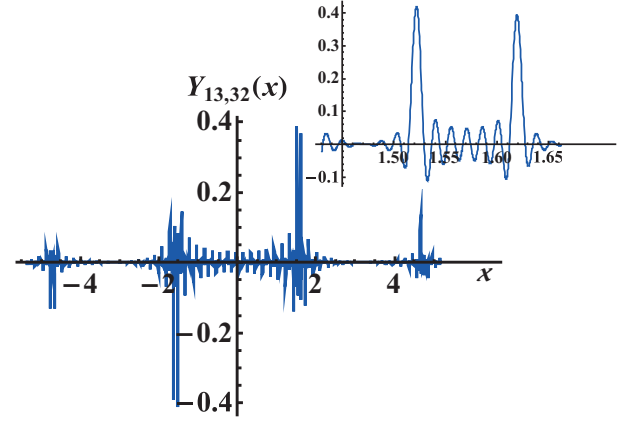


FIG. 7: Plot of the function $Y_{M,N}(x)$ for the parameters for simultaneous QPM of four-photon down conversion. The number of domains is $N = 32$ and that of the blocks is $M = 13$. The insert is detailed the structure of twin-peaks.

reversed crystal mismatch vectors are $\Delta k_1 = 1,56\mu m^{-1}$, $\Delta k_1 = -1,312\mu m^{-1}$, these parameters are taken from the experimental results for fourth-harmonic generation [18]. We found the parameters of the phase-reversed structure for simultaneous realization of the four-photon down-conversion. QPM of mentioned process is realized and perfectly matched with the experimental results for parameters $l = 2, 2\mu m$, $M = 13$ and $N = 32$ (see, Fig. 7).

IV. CONCLUSION

In conclusion, we have analyzed phase-reversed QPM structures in superlattice as the ensemble of domains and blocks of second-order nonlinear layers. In the result, the effective nonlinear coefficient $G(\Delta k)$ of three-wave interaction has been obtained as the product of the standard phase-matching spectral function for a single segment and the assemble function. Generally used phenomenological method for description of multilayered structures required huge number of domains and blocks to provide the periodicity of the function. Our investigations shows that the detailed method we have used for description of the systems qualitatively matches with phenomenological results for the big number of domains N and blocks M , moreover, it is applicable for the finite number of layers. The results shows that we can QPM various multiphoton processes managing number of domains and blocks. We have demonstrated that double phase-matched interaction on the base of phase-reversed QPM structures in superlattice becomes possible in a wide region of the optical wavelengths, particularly, for production of muliphoton states.

Acknowledgments

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